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Incentives for implementing new pollution-control technologies are both regulatory and economic. Given considerable regulatory pressure, e.g. , the promulgation of a NESHAPS for NOx emissions in CY 2000, new de-NOx technologies are being explored. This project is currently evaluating non-thermal plasma (NTP) technologies for treating jet-engine exhaust and other hazardous air pollutants. To meet a project milestone this White Paper will present our initial design options for NTP reactor systems for a field-pilot demonstratiion on Cruise Missile Test Cell (CMTC) exhaust at Tinker AFB. The field-pilot demonstration is necessary to provide further data and operating experience to more fully evaluate economic and performance projections for NTP de-NOx technology and to design larger systems with confidence. From the design options presented here, we will downselect the set to 2 treatment systems and consider fielding both. If the budget is not sufficient for 2 reactor systems, only one will be fielded. This paper will discuss the exhaust stream to be addressed, the test setup, the candidate reactor systems, and projected operating parameters and specifications for the field-pilot units Because the cost and logistics of using an electron-beam NTP reactor are, respectively, too high and too complicated for this project, we have limited our candidate systems to those based on electric-discharge-driven NTP reactors (which previous economic analyses have shown to be more cost effective).

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Initial Designs of Electric-Discharge Non-Thermal Plasma Field-Pilot Demonstration Units for NO_x Removal in Jet-Engine Exhaust: White Paper for SERDP Project CP-1038

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Abstract

Incentives for implementing new pollution-control technologies are both regulatory and economic. Given considerable regulatory pressure, e.g., the promulgation of a NESHAPS (National Emissions Standard for Hazardous Air Pollutants) for NO_x emissions in CY 2000, new de-NO_x technologies are being explored. This project is currently evaluating non-thermal plasma (NTP) technologies for treating jet-engine exhaust and other hazardous air pollutants. To meet a project milestone (*viz.,* document initial field-pilot designs), this White Paper will present our initial design options for NTP reactor systems for a field-pilot demonstration on Cruise Missile Test Cell (CMTC) exhaust at Tinker Air Force Base (currently scheduled for September 1999). The field-pilot demonstration is necessary to provide further data and operating experience to more fully evaluate economic and performance projections for NTP de-NO_x technology and to design larger systems with confidence. From the design options presented here, we will downselect the set to two treatment systems and consider fielding both, if the projected costs of fabrication and demonstration fit our project budget. If the budget is not sufficient for two reactor systems, only one will be fielded. This paper will discuss the exhaust stream to be addressed, the test setup, the candidate reactor systems, and projected operating parameters and specifications for the field-pilot units. Because the cost and logistics of using an electron-beam NTP reactor are, respectively, too high and too complicated for this project, we have limited our candidate systems to those based on electric-discharge-driven NTP reactors (which previous economic analyses have shown to be more cost effective).

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- ***Hybrid NTP Reactor-Adsorber System***
- ***Corona Radical Shower Plasma Reactor System***

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Introduction (Purpose of White Paper and Main Topics Covered)

The purpose of SERDP project CP-1038 is to evaluate and develop non-thermal plasma (NTP) reactor technology for Department of Defense (DoD) air emissions control applications. The primary focus is on oxides of nitrogen (NO_x) and a secondary focus on hazardous air pollutants (HAPs), especially volatile organic compounds (VOCs). Example NO_x sources are jet engine test cells (JETCs) and diesel-engine powered electrical generators. Example VOCs are organic solvents used in painting, paint-stripping, and parts cleaning.

A project milestone for FY99 is to document initial designs for NTP reactor systems that are candidates for a field-pilot demonstration on the treatment of jet-engine exhaust [1]. This White Paper will offer four candidate system designs based on electric-discharge-driven NTP reactor systems for emissions control (given the scope and budget for Project CP-1038, we do not consider an electron-beam system to be a field-pilot demonstration candidate for this work). The exhaust stream to be addressed, the test setup, salient features of the design of the candidate systems, and the projected operating specifications for the systems will be presented.

Exhaust Stream to be Addressed

There are several studies and reports that address jet-engine emissions arising from engine test facilities (Spicer et al 1988 [2], 1990 [3]; Walker 1996 [4]). Representative emissions of the major compounds of concern are shown in Table 1 for F101 and F110 jet engines operated in jet engine test cells (JETCs) at Tinker Air Force Base, Oklahoma [3].

Table 1: Measured emissions for Tinker AFB JETCs (F101 & F110 engines) [3].

Power setting	Test No.	THC (ppmC)	NO_x (ppm)	NO (ppm)	CO (ppm)	CO_2 (%)
F101 Engine						
Idle	TAFB-1-6-17	6.5	6.9	5.0	50.0	0.50
44%	TAFB-2-6-17	3.5	28.5	25.5	8.0	0.98
75%	TAFB-3-6-17	2.5	68.0	62.0	8.0	1.52
Intermediate	TAFB-4-6-17	3.0	140.0	133.0	11.0	2.02
Augmentation (Stage 1)*	TAFB-5-6-17	287.0	21.8	7.2	110.0	0.32
F110 Engine						
Idle	TAFB-1-6-15	7.0	13.8	11.2	85.0	0.98
30%	TAFB-2-6-15	6.0	30.0	28.0	23.0	1.25
63%	TAFB-3-6-15	3.0	97.0	92.0	13.0	2.35
Intermediate	TAFB-4-6-15	3.5	243.0	227.0	15.0	3.17
Augmentation (Stage 1)*	TAFB-5-6-15	335.0	21.5	3.7	178.0	0.41
Intermediate (Rooftop)*	------	< 7.0	26.0	25.0	6.0	0.28

* Measurements made with ~ 20-50:1 diluted exhaust.

It should be noted that the exhaust streams form JETCs and CMTCs are diluted by factors in the range of 10-50 with air added at the engine exhaust duct/augmentor intakes. Typically, this reduces the concentration of pollutants (e.g., to 10s ppm NO_x and hydrocarbons) but increases the overall exhaust-gas flow discharged to the atmosphere.

Table 2 shows a summary emissions inventory for Tinker AFB JETCs 1-12 for the year 1995; when 3,414,836 gallons of JP-5 fuel were consumed in a time period of 4420 hours of operation [4]. The emissions were calculated on the basis of fuel consumption but not directly measured.

Table 2: Calculated emissions inventory for twelve JETCs at Tinker AFB for CY1995.

Compound	Emission Inventory (ton/yr)
NO_x	113.01
SO_x	30.71
Aggregate hydrocarbons	100.45
CO	156.34
Particulates	26.72
PM-10	4.45

Based on the data taken from Spicer 1990 [3] and Walker 1996 [4], a model emissions profile for a representative JETC can be defined. However, our plans for a field-pilot demonstration for this project call for testing NTP jet-engine emissions treatment on a Cruise Missile Test Cell (CMTC) at Tinker AFB (which employs F107 and F112 engines). In contrast to the Tinker JETCs, the actual emissions from the CMTCs have not been characterized. Therefore, our approach is to: 1) work with Tinker to have the emissions characterized for a CMTC; and 2) formulate a model emissions profile, based on the measured and calculated profiles for JETCs. These items will be used in setting the final operating parameters for the field-pilot equipment and in making cost-analysis and economic projections for the treatment of jet-engine emissions by NTP systems and in making comparisons with conventional flue-gas treatment technologies.

NTP technology probably has applications for treating air emissions from other sources of interest to the DoD; e.g., industrial boilers and furnaces; Aerospace Ground Equipment (AGE), including diesel-powered electrical generators, compressors, hydraulic test stands, and weapons loading units; and emergency electrical power generators. Means of calculating inventories for air pollutants arising from such sources have been formulated and documented by Jagielski et al 1994 [5].

The exhaust from JETCs has been extensively characterized by Spicer et al. However, the CMTC emissions have not been characterized. In the absence of exhaust-gas sampling and characterization, a CMTC will be assumed to have similar concentrations of emitted pollutants. But the gas flows, both that coming directly out of the engine and the diluted flow, have lower flow rates because the CMTC engines are smaller than those normally employed in JETCs. Before actual field tests, Tinker AFB will arrange for sampling and characterization of the CMTC emissions. Table 3 lists the exhaust-gas parameters that we will use for initial designs of the field-pilot equipment.

Table 3: Expected exhaust-gas data and conditions.

Source Data	Variable	Units	JETC Value	CMTC Value
Gas Flow	Q_{gas}	Nm^3/h	1.0E+05 1.7E+06	6.6E+03 1.3E+04
Fuel			JP-5	JP-5
Final Exhaust-Gas Composition				
N_2	C_{N2}	%	80.98	80.98
O_2	C_{O2}	%	18.00	18.00
CO_2	C_{CO2}	%	0.50	0.50
H_2O	C_{H2O}	%	0.50	0.50
Density (Normal)	D_{gas}	kg/Nm^3	1.283	1.283
Exhaust Gas Temperature	T_{gas}	C	25	25
NTP Inlet Temperature	T_{NTPin}	C	25	25
Pressure	Pr_{gas}	mm Hg	720	720
Emission Data				
NO_X	C_{NOx}	ppm	36.00	36.00
SO_2	C_{SO2}	ppm	4.59	4.59
HC (VOC)	C_{HC}	ppm	60.00	60.00
CO	C_{CO}	ppm	53.36	53.36
Particles	C_{part}	mg/Nm^3	-	-
	Variable	**Units**	**Value**	
NH_3 Stoichiometric Ratio to NO and SO_2: 1.5 for both JETCs and CMTCs				

Test Setup

Under an agreement with Tinker AFB in Oklahoma City, we plan to test one or more NTP reactor system concepts on jet-engine exhaust. Because of size and operational flexibility considerations, we envision using a CMTC for conducting a field-pilot demonstration. Figure 1 below shows a schematic diagram of the planned test setup. In this arrangement, the diluted exhaust will be treated because it is the emissions stream that is actually discharged to the atmosphere. A sampling blower or pump will be used to draw exhaust gas into the test reactor system. Because we will be treating only a slipstream of 100-500 SCFM (59-294 Nm^3/h) capacity, there will be no deleterious back-pressure effects on the engine. The on-line analysis equipment is expected to consist of combustion gas analyzers (CGAs) and, possibly a gas chromatograph - mass spectrometer (GC-MS). Additionally, analysis of byproduct liquid and particulate effluents are envisioned to be performed through other, off-line analytical techniques.

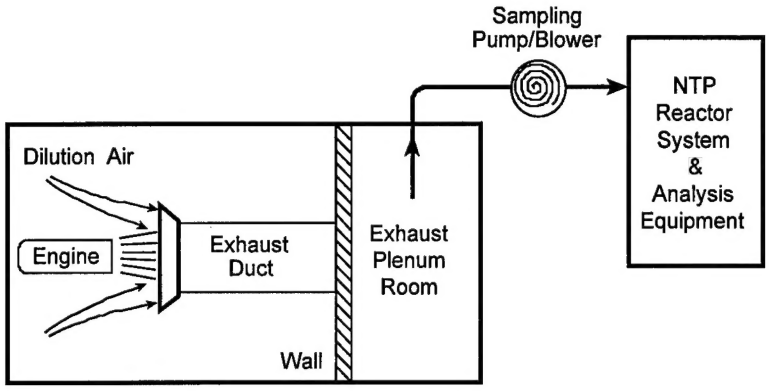

Figure 1: Schematic diagram of NTP reactor system setup for field-pilot demonstration on a CMTC.

Candidate Designs for Field-Pilot Demonstration Equipment

We have chosen four initial candidate NTP reactor systems (all based on electric discharges) for the field-pilot demonstration:

- •Pulsed Corona Plasma Reactor System
- •Dielectric-Barrier (Silent Discharge) Plasma Reactor System
- •Hybrid/NTP Reactor-Adsorber System
- •Corona Radical Shower Plasma Reactor System.

Because the cost and logistics of using an electron-beam NTP reactor are, respectively, too high and too complicated for this project, we have limited our candidate systems to those based on electric-discharge-driven NTP reactors (which previous economic analyses have shown to be more cost effective, Kim & Chang 1998 [6], Rosocha et al 1998 [7]).

Active species/radicals capable of decomposing NO_x, SO_x, and hydrocarbons are created in all four of the above-mentioned reactors. However, some have particular advantages. Each type of NTP reactor system will be discussed in greater detail further below.

As a rough estimate, we assume that the production of radicals is the same for all four reactors listed above (which is not explicitly true) so that we can provide an estimate of the required plasma power for a given removal fraction of NO. The specific plasma energy for a one e-fold removal (~ 63%) of NO - the figure for which we are planning - is approximately 50 J/L in electric discharge reactors. The average plasma power requirement is calculated form the equation

$$P = \bar{E}\, Q ,$$

where P is the power, $\bar{E}$ is the plasma energy density (50 J/L for our case), and Q is the gas flow rate through the reactor. For 100 and 500 SCFM (59 and 294 Nm^3/h) gas flow rates, the average plasma powers are 2.4 kW and 11.8 kW, respectively.

Depending on the exhaust-gas conditions (e.g., humidity and hydrocarbon content) and the potential use of additives, the overall de-NO_x removal chemistry in our candidate NTP reactor systems can be either oxidative or reductive. This will influence the final byproduct effluent distributions. In the oxidative case under humid conditions, the most prevalent byproduct is nitric

acid (HNO_3), which dictates the use of base scrubbers to neutralize the acid. Under dry conditions, the formation of NO_2 is favored. However NO_2 is more easily adsorbed by activated carbon and can be captured and subjected to further treatment. With additives such as ammonia (NH_3), methane (CH_4), ethylene (C_2H_4), or part of the actual exhaust gas, the product distribution can be shifted to particles (e.g., useful agricultural fertilizer like ammonium nitrate - NH_4NO_3) which can be collected with an electrostatic precipitator.

Pulsed Corona Plasma Reactor System

A generic pulsed or DC corona reactor is shown in Figure 2 below. In pulsed corona, the combination of a fine wire and a short, high-voltage pulse provides a gas breakdown electric field that is enhanced over the normal DC breakdown field. Several wires can be used to provide a larger active gas-treatment volume.

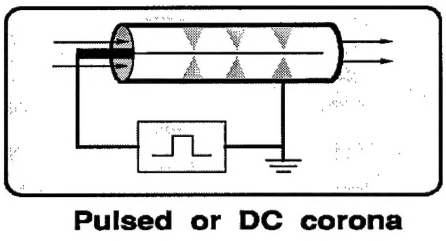

Figure 2: Schematic diagram of a generic wire-tube pulsed or DC corona NTP reactor.

A schematic diagram illustrating a more specific pulsed corona reactor system is shown in Figure 3. In this system, ammonia (NH_3) or ethylene (C_2H_2) can be supplied as additives to enhance the production of useful reactive species. Similarly, a small portion of the actual engine exhaust can be injected at the reactor intake to use hydrocarbons entrained in the exhaust gas for active-species enhancement.

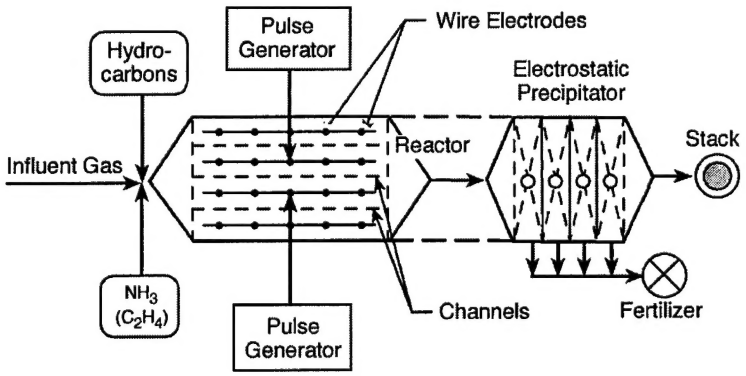

Figure 3: Schematic diagram of pulsed corona reactor system for exhaust-gas emission treatment, including electrostatic precipitator for particulate collection.

Dielectric-Barrier Discharge Plasma Reactor System

A generic dielectric-barrier (silent discharge) reactor is shown in Figure 4 below. In an AC-driven barrier discharge, the buildup of charge on the dielectric automatically terminates the microdischarge streamers, thus producing a short, electron-energetic pulse and eliminating the need for more expensive and/or more complicated pulsed power supplies.

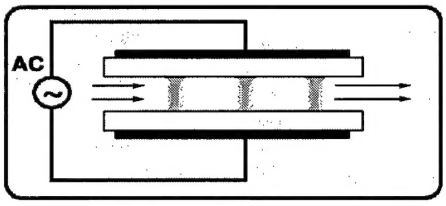

Figure 4: Simple schematic diagram for a dielectric-barrier discharge reactor.

At Los Alamos, the dielectric-barrier reactor has been extensively studied for the decomposition of VOCs (especially chlorinated hydrocarbons) and four field-pilot demonstrations have been carried out with modular reactors. Reactor banks with average plasma power as much as 10 kW have been employed in such tests (Rosocha 1997 [8]). Commercialization of the technology for VOC/air toxics treatment under specific fields of use is now in progress with a commercial partner. Figure 5 shows an illustration of a mobile unit that was employed at McClellan AFB in an innovative-technology demonstration on treating multiple VOCs and other chemicals entrained in the soil. Similar units were also employed at the DOE Savannah River Site for treating VOCs entrained in soil and groundwater [9] and at Tinker AFB for treating low-concentration VOCs extracted from groundwater [10].

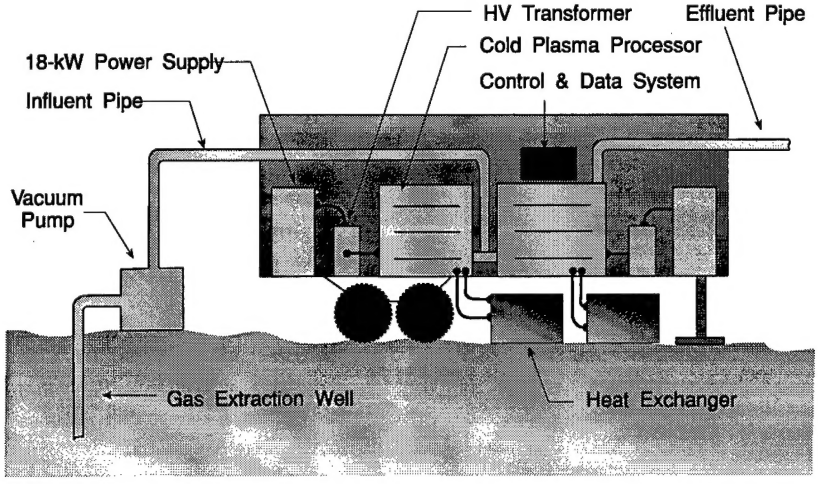

Figure 5: Illustration of mobile dielectric-barrier NTP reactor system employed for VOC decomposition tests at McClellan AFB. Each plasma reactor tank operated at up to 10 kW of plasma power.

An example of an NTP reactor that has been commercialized for flue-gas treatment is the Tecolytic™ modified dielectric-barrier reactor + lime scrubber system (from Thermo-Power Corp., Bittenson & Breault 1998 [11]). The company's stated objective is to "develop a zero discharge NO_x control process using no hazardous reagents or catalysts". Figure 6 shows a schematic diagram of the system. The NTP reactor consists of a housing to hold a large array of metal rods covered by ceramic-dielectric insulators and to hold the associated high-voltage insulated feedthroughs. The rods are essentially arranged such that a high-voltage electrode is surrounded by four grounded nearest-neighbor rods. The high-voltage "corona" rods are connected to a HV/AC power supply to supply the necessary voltage and current to produce an electrical discharge in the gas space between the rods. Flue gas is flown across the electrodes, entering the reactor housing at one end and exiting the opposite end. The NTP-treated gas is then sent to a wet scrubber, using $Mg(OH)_2$ and slaked Mg-enhanced lime, which scrubs out SO_2 to make gypsum ($CaSO_4 \cdot 2H_2O$), which is a salable commodity. In humid flue gas, much of the NO_x is converted in nitrate products (e.g., acids which can be neutralized or collected as products). Clean effluent gas is vented to the atmosphere.

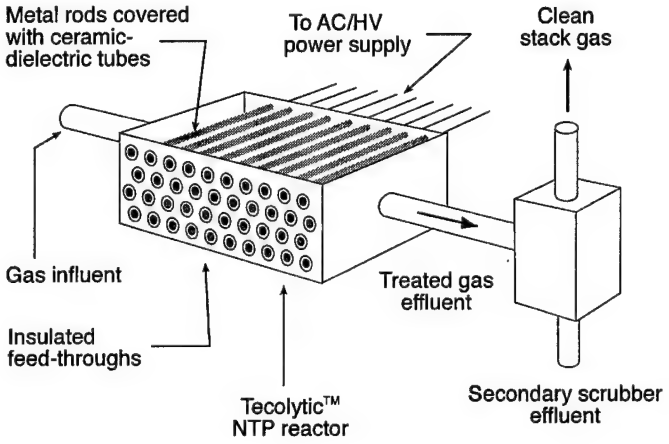

Figure 6: Schematic diagram of commercial Tecolytic™ modified dielectrc-barrier NTP reactor system for de-NO_X/SO_X (flue-gas treatment).

Within the past few months, one of these systems has been installed on the Miami Fort power-plant facility and field tests are in progress. Data from these tests will be highly useful in establishing further benchmarks for the economic model and in lending further credence for the acceptance of NTP technology as an alternative to conventional de-NO_x methods.

Hybrid NTP Reactor-Adsorber Systems

We define a hybrid NTP emissions-control system as a combination of one or more NTP reactors with an adsorber, a catalyst, or another NTP reactor. Our interest in hybrid systems arises from the fact: if the operating lifetime and/or effectiveness of GAC can be improved, the treatment costs will decrease.

There are two simple ways to combine an NTP stage with a GAC stage: place the NTP stage in series with the GAC stage, thus lessening the load on the GAC; or place the NTP stage in

parallel with the GAC stage and use it to regenerate the GAC under more favorable conditions than the heat/steam regeneration methods typically employed. The expected advantages of such an NTP-GAC hybrid system are:

- Prolonged life of GAC filters (with an associated operating cost reduction)
- Application to a broader range of exhaust-gas flow rates, types of pollutants, and pollutant concentrations
- Potential for reducing the dependence of treatment cost on pollutant concentration
- Pollutants are destroyed by the NTP stage, rather than simply captured
- NTP system can incorporate feedback to aid in optimizing the treatment efficiency and costs.

For many applications, end-of-pipe emissions treatment is the norm. However, one can also conceive of restricting the treatment closer to the point-of-use, or integrating the emissions treatment equipment directly into the process which produces the emissions. For the purposes of this report, an end-of-pipe application will sufficiently illustrate the hybrid system concept.

Serial-Mode NTP Reactor Hybrid Architecture

In a serial-mode hybrid system, an NTP reactor precedes an adsorber bank (see Figure 7). As mentioned earlier, adsorbers such as activated carbon, are commonly-employed but cost-intensive treatment methods (mainly because of regeneration, reactivation, or disposal costs). For the serial-mode hybrid, the load on the adsorber stage can be possibly reduced by 50-75% by the pretreatment action of the NTP reactor. This results in a significant change in the overall treatment economics because the useful adsorber lifetime can be greatly increased, while the NTP reactor does not have to operate in an energy-demanding, high-removal regime (the energy cost per pollutant molecule destroyed is a logarithmic function of the degree of removal). Additionally, one can envision tailoring the adsorber to better match the compounds which the NTP reactor produces, thereby increasing the overall process effectiveness. That is, one is not necessarily constrained to the use of GAC - superior adsorbents are most likely available and adsorber technology is expected to advance in the future.

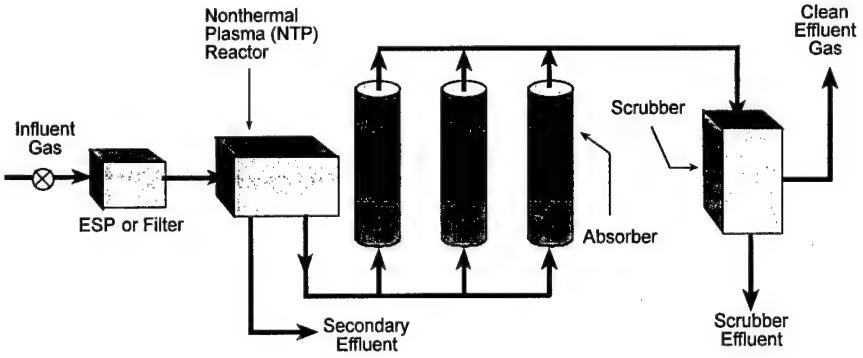

Figure 7: Serial-mode NTP-absorber architecture. Disposal and/or regeneration economic advantage comes from reducing the load on the absorber or converting the pollutants to more easily-absorbed compounds.

Regenerative-Mode NTP Reactor Hybrid Architecture

In a regenerative-mode (or 'trap and treat') hybrid system, an NTP reactor is used to regenerate a pollutant-adsorber bank (see Figure 8). Here the adsorber traps the pollutants (NO_x or VOCs) while operating at a high off-gas flow rate, but is regenerated off-line at more economical conditions. Such conditions can be a lower flow rate and, hence, a lower power demand (and associated lower power cost when operating at electrical utility off-peak times). This architecture is particularly attractive for episodic emissions (e.g., JETCs and CMTCs), where high-flow operation and regeneration can be easily divided into separate functions.

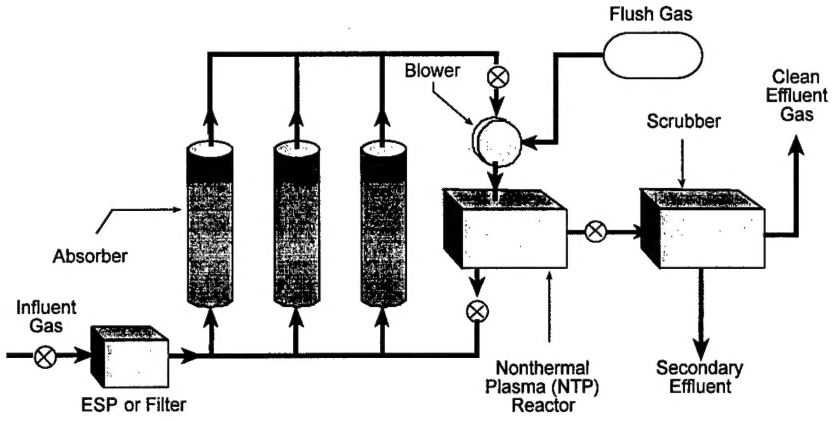

Figure 8: Regeneration-mode NTP-absorber architecture. Economic and performance advantages may be gained by regenerating the absorbers off-line from pollutant capture, but employing on-site, rather than off-site handling.

Two additional key advantages of the NTP regenerative hybrid are: the ability to incorporate electronic feedback into the process, thereby operating the system at more optimal treatment conditions and costs; and the ability to flush the adsorbent with a tailored gas mixture, thereby more effectively controlling the destruction chemistry, the formation of undesirable byproducts, and the overall effectiveness and treatment costs.

Corona Radical Shower Plasma Reactor System

There are several ways to combine one or more NTP reactors with other NTP stages. A promising, novel corona reactor called the Corona Radical Shower (CRS) or radical injector, that employs a small NTP reactor to inject beneficial active species into the main NTP reactor, has been demonstrated by Kanazawa et al 1997 [12] and Chang et al 1998 [13]. This device is more fully described in an earlier report to SERDP (Matsuoka et al 1997 [14]). Here, a brief summary of the system will be presented.

In the CRS system (see Figure 9), arrays of small nozzles or showers, each with a small bleed-gas flow, are introduced into a wire-plate DC corona reactor. The purpose of the nozzles is to create desirable active species and inject them into the larger main corona treatment region, which enhances the overall pollutant-removal effectiveness. The injected active species can be tailored to the particular pollutant stream being treated by selecting the shower-injector bleed-gas so that it produces active species that are particularly effective in decomposing the target pollutant.

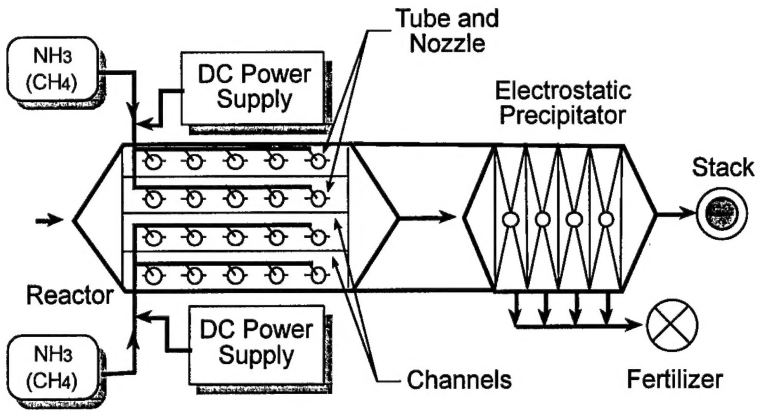

Figure 9: Schematic diagram of CRS reactor. Ammonia (NH_3) or methane (CH_4) are added to generate radicals that drive reactions leading to the formation of particulates; these particulates are then captured by the electrostatic precipitator. Some of the captured products are useful for agricultural fertilizer (e.g., ammonium nitrate, NH_4NO_3).

Experiments by our collaborators at McMaster University have shown that, for NO removal, ammonia (NH_3) or a hydrocarbon like methane (CH_4) or ethylene (C_2H_4) are useful injector-gas additives. It is interesting to note that McMaster has also shown that, for JETC de-NO_x, normally-present hydrocarbons in the exhaust stream can enhance the de-NO_x process. In this case, air or a slipstream of the JETC exhaust itself is effective as a shower-injector gas, without requiring additional external additives (like NH_3). The economic advantages of the CRS system have been described previously [6, 7].

Projected Operating Specifications

The operating specifications and conditions for the field-pilot demonstration equipment have been projected from lab-scale work, pilot-plant data taken from the literature, and from our own experience. Because of the scope of this project, we will limit the field-pilot equipment to a slipstream capacity, but of sufficient gas flow to have confidence in further extrapolation to full-scale systems. Table 4 below summarizes the initial projected operating specifications for the candidate NTP de-NO_x reactor systems to be tested on a CMTC. These specifications will be refined and expanded as our downselection and detailed design processes progress.

Table 4: Initial specifications for candidate demonstration NTP reactors for CMTC tests.

Parameters	Exhaust-Gas Flow Rate		Other
Pollutant Concentrations	100 SCFM (59 Nm^3/h)	500 SCFM (294 Nm^3/h)	Removal Goal (Both Flow Rates)
[NO_x], ppm	36.00	36.00	63%
[SO_2], ppm	4.59	4.59	63%
[HC (VOC)], ppm	60.00	60.00	63%
[CO], ppm	53.36	53.36	0.00
[Particles], mg/Nm^3	-	-	-
Plasma Specific Energy	50 J/L	50 J/L	
Plasma Power (Approximate)	2.4 kW	11.8 kW	
Operating Pressure	~ 1 local atm	~ 1 local atm	
Potential Additives	NH_3, CH_4, C_2H_4, exhaust	NH_3, CH_4, C_2H_4, exhaust	
NH_3 Stoichiometric Ratio to NO and SO_2	1.5	1.5	

Conclusions/Summary

In this report, we have presented four candidate designs for a demonstration field-pilot NTP jet-engine exhaust de-NO_x reactor system. Realizing the performance and economic shortcomings of stand-alone NTP reactors, some workers in this discipline (particularly this SERDP project team) have proposed the use of staged or hybrid systems to better match particular air-emissions control applications. Initial evaluations of hybrids show promising performance and economics. However, rigorous pilot-plant tests are required to provide further data and operating experience to more fully evaluate economic and performance projections and to extrapolate designs to full-scale units. The demonstration of a small-scale, field-pilot unit directed toward scale-up is a key goal of this project and a key goal in providing the DoD with further information to provide a basis for selecting the most appropriate NTP technology for a given emissions-control application.

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